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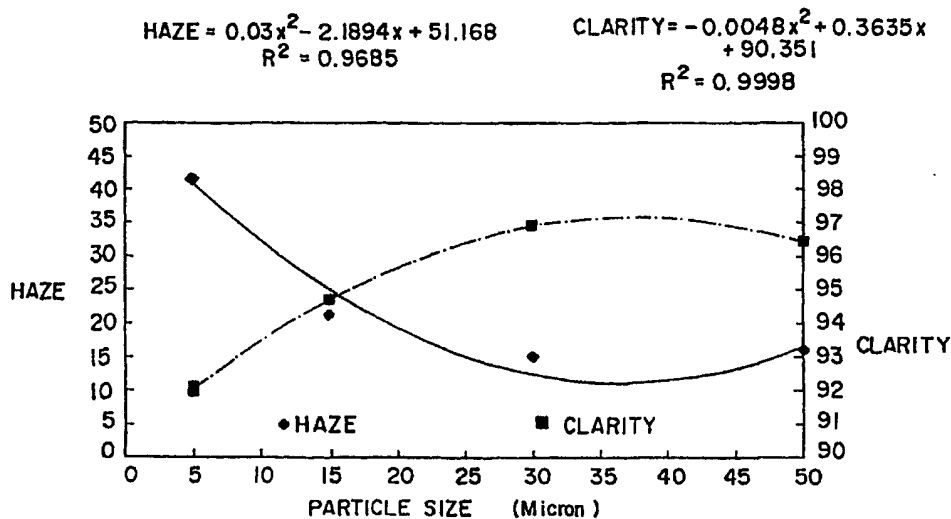
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(54) Title: THERMOPLASTIC ARTICLE HAVING LOW CLARITY AND LOW HAZE



(57) Abstract

There is described herein a thermoplastic article which has a high transmittance, less than about 95% haze and low clarity. If this article is placed in front of an object, the observer on the far side of the article will see the object, but not clearly.

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**THERMOPLASTIC ARTICLE HAVING LOW CLARITY AND LOW HAZE**BACKGROUND

This application relates generally to translucent thermoplastic articles. Specifically, this application relates to thermoplastic articles having relatively high percent light transmission, haze less than about 95%, and relatively low percent clarity. For purposes of this application, levels of haze which may be  
5 high in an absolute sense, but are still between 70 and 95 %, will be referred to as "relatively low haze." If an observer looks through such an article at an object behind the article, the observer can see that there is a particular object a distance behind the article (relatively low haze), but the observer can not clearly see the object (low clarity).

10 Various efforts have been made to make translucent thermoplastic articles having unique visual appearances. For example, thermoplastic articles having very high haze (above about 95%) have been made by incorporating particles of a polymer into a polymer matrix having a different refractive index. Specifically, Japanese Patent No. 3,143,950 discloses light  
15 fixture covers made by dispersing polymer particles in a polymer matrix having a different refractive index. Such materials have been useful for enclosures where the intent is not to let the observer see any apparatus behind the article. Very high haze, low clarity light fixture enclosures are made from this material because the emitted light is well dispersed across the entire  
20 enclosure. Also, the light bulb remains hidden behind the enclosure when it is not lit.

Typically, articles having relatively low haze also have high clarity. For example, polycarbonate compositions comprising ZnO as a light diffuser typically have clarity above 90 % when haze is near 90%. According to ASTM

D 1003, which is hereby incorporated by reference, haze is defined as the percentage of transmitted light that deviates from the incident beam by more than  $2.5^\circ$  on average. Clarity is defined as the percentage of transmitted light that deviates from the incident by more than 0, but less than  $2.5^\circ$  on average.

5 Conventional materials which have relatively low haze (i.e., a smaller degree of  $> 2.5^\circ$  scattering) also have high clarity (i.e., a smaller degree of  $< 2.5^\circ$  scattering). This relationship holds true even when the absolute level of haze is high. The "relatively low" haze level is less than about 95%.

For some applications, it is desirable to achieve a translucent material having relatively low haze and relatively low clarity. For example, in applications such as privacy windows, and more recently business equipment housings, it is desirable to have high light transmission, relatively low haze, and relatively low clarity so that objects can be seen behind the material, but not clearly.

10

There have been previous attempts to make translucent polycarbonate resin compositions by incorporating zinc oxide into polycarbonate. There are several disadvantages to compositions which incorporate zinc oxide or other inorganic light diffusing agents. Specifically, these agents tend to react with polycarbonate and other thermoplastics to cause degradation of the physical characteristics of the thermoplastic. Additionally, compositions comprising zinc oxide tend to have very high clarity at relatively low levels of haze which are still high in an absolute sense (e.g., 40% to about 98%).

15  
20

#### SUMMARY OF THE INVENTION

There is described herein a thermoplastic article which has a high percent light transmission, relatively low haze and low clarity. If this article is placed in front of an object, the observer on the far side of the article will see the object, but not clearly. In a preferred embodiment of the invention, the

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thermoplastic article will have a percentage light transmission above 60%, a haze value between 30 and 95%, and a clarity of above 70%, but below 95%. In a more preferred embodiment of the invention, the clarity will be below 85%.

5           It has been found that these limits on haze and clarity may be achieved if the size of the light diffuser spherical thermoplastic particles is carefully controlled. Specifically, when the size of the transparent thermoplastic particles is reduced below a certain point, the clarity begins to decline and the haze begins to increase. Above this critical size limit, there is little  
10           dependence of clarity and haze on particle size, and the clarity will be too high to give the desired optical effect -- a slight blurring of objects seen through the material. By carefully selecting a particle size below the critical size limit, one can obtain the desired properties of relatively low haze, and low clarity.

15           The article described herein comprises roughly spherical transparent thermoplastic particles dispersed in a transparent matrix thermoplastic resin. The particles and the matrix thermoplastic resin have differing refractive indices. When the size, loading and type of spherical transparent thermoplastic particles is chosen appropriately, a material will be obtained  
20           which has a relatively low clarity without sufficient haze to prevent an observer from discerning objects behind the article.

          The article is preferably substantially free from inorganic light diffusion agents, and thereby avoids degradation caused by such additives.

          Other features, aspects and advantages of the invention will become  
25           better understood with reference to the following detailed description of some preferred embodiments of the invention, the drawings and the appended claims.

### BRIEF DESCRIPTION OF THE DRAWINGS

FIG. 1 is a plot of percent light transmission versus sample number for the six samples described in Example 1.

FIG. 2 is a plot of percent haze versus sample number for the six samples described in Example 1.

5           FIG. 3 is a plot of percent clarity versus sample number for the six samples described in Example 1.

FIG. 4 is a plot of percent clarity and haze versus particle size for a polycarbonate/PMMA article as described in Example 2. (PMMA is polymethylmethacrylate).

10          FIG. 5 is a plot of haze versus PMMA particle size for samples having various thicknesses as described in Example 2.

FIG. 6 is a plot of clarity versus PMMA particle size for samples having various thicknesses as described in Example 2.

### DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENTS

15           A typical thermoplastic article according to the invention comprises a matrix of transparent matrix thermoplastic resin (hereinafter "the matrix") and transparent spherical thermoplastic particles (hereinafter "spherical particles") suspended therein. Such articles have a percent light transmission above 60%, a haze of less than 95%, and a clarity of greater than 70% but less than 97%. To obtain these optical characteristics the particles must be selected  
20           carefully. Specifically, they must have an average diameter small enough such that the clarity is below its maximum versus the average diameter of the spherical particles (i.e., as shown in Figures 4 and 6, the clarity remains very flat above a certain diameter threshold, but begins to taper off steeply below

this point). In a preferred embodiment of the invention, the spherical particles are sufficiently small such that clarity is less than 85%. At the same time, the spherical particle size can not be made too small because the haze varies inversely with particle diameter. Preferably, the spherical particles have an average diameter large enough such that the haze is above its minimum value versus particle size, but below 95%. The percent light transmission is preferably maintained above 60%.

In a more preferred embodiment of the invention wherein the matrix is polycarbonate and the spherical particles are PMMA particles, the percent light transmission is above 85%, the haze is less than 95%, and the clarity is less than 85%. In this embodiment, the loading of PMMA particles is preferably from 0.01 to 0.50, and more preferably 0.05 to 0.50 parts per hundred resin based on the matrix.

The transparent thermoplastic matrix described above may be any transparent thermoplastic material which is compatible with PMMA spherical light diffusing agents. Preferred matrix materials include polycarbonates, polyetherimides, transparent polyimides, transparent polyamides (nylons), polyesters such as transparent aliphatic polyesters (e.g., polycyclohexane carboxylic acid), transparent polycarbonate-polyester blends, polysulfones, polyether and polyphenyl sulfones, styrene acrylonitrile (SAN), polyethylene polystyrene, and miscible transparent polystyrene-polyphenylene oxide (PS-PPO) blends, acrylics, polycarbonate-polysiloxanes, polyetherimide-polysiloxanes, polyarylates (e.g., isophthalate resorcinol terephthalate), and blends and copolymers of all of the above. More preferred transparent thermoplastic matrix materials are polycarbonate homopolymers or copolymers, polyester carbonates and polyethylene terephthalate (PET). The most preferred matrix material is an aromatic polycarbonate homopolymer based primarily on the bisphenol-A monomer. The synthesis of such materials is well known in the art. For example, U.S. Patent No. 5,364,926

describes the melt process for making polycarbonate, and is incorporated by reference herein. The interfacial and solid state process can also be used. Additionally, it is possible to use matrix materials that are not perfectly transparent, provided they are sufficiently transparent to achieve the visual  
5 effect described above.

The spherical particles may be any transparent thermoplastic polymer or other light diffuser which has a refractive index (hereinafter "R.I.") different from that of the matrix. Preferably, the R.I. of the light diffuser differs from that of the matrix by at least 0.01. Suitable light diffusers  
10 included polytetrafluoroethylene, zinc oxide, and PMMA. Among these, Techpolymer MBX-series crosslinked PMMA microspheres, which are available in various diameters from Nagase America (e.g., 5 -50 micron avg. diameter), are preferred. In a more preferred embodiment of the invention, the matrix thermoplastic resin is polycarbonate having an R.I. of 1.56 to 1.62,  
15 and the spherical particles are PMMA having an R.I. of 1.46 to 1.53.

In a preferred embodiment of the invention, the spherical transparent thermoplastic particles have an average diameter which is sufficiently small such that the clarity of the article is below its maximum value with respect to the average diameter of the spheres.

20 In cases where the matrix thermoplastic resin is polycarbonate and the spherical particles are PMMA, this condition is typically satisfied when the average particle diameter is less than about 30 microns (see FIG. 4). In such cases, the particle size is more preferably below 15 microns, and most preferably between 3 and 10 microns. In an alternate embodiment of the  
25 invention, there may be two or more sets of PMMA particles wherein at least one set has a particle size below 30 microns. Preferred PMMA particles for incorporation in a polycarbonate matrix have a specific gravity of about 1.10 to 1.30, and are highly crosslinked (e.g., essentially 100% crosslinked).



In a more preferred embodiment of the invention the diameter of the spherical particles is sufficiently small such that the clarity is less than 0.95 of its maximum value versus the diameter of the spherical particles. In a most preferred embodiment, the spherical particles are sufficiently such that the clarity is less than 0.93 of its maximum value versus the diameter of the particles.

PMMA spherical particles in the various sizes between 1 and 50 micron diameter are commercially available from Nagase America. Methods of making such particles are known, and are described, for example, in Japanese Published Patent Application No. JP 6220290, which is incorporated herein by reference.

The thermoplastic matrix may optionally further contain an optical brightening agent, additional pigments and/or a fluorescent dye. Adding an optical brightening agent helps produce a brighter color for the article. Suitable optical brightening agents include aromatic stilbene derivatives, aromatic benzoxazole derivatives, or aromatic stilbene benzoxazole derivatives. Among these optical brightening agents, Uvitex OB from Ciba Specialty Chemicals [2,5-bis(5'-tert-butyl-2-benzoxazolyl)thiophene] is preferred.

Adding a fluorescent dyestuff generates striking visual effects for the article. Suitable fluorescent dyestuffs include Permanent Pink R (Color Index Pigment Red 181, from Clariant Corporation), Hostasol Red 5B (Color Index #73300, CAS # 522-75-8, from Clariant Corporation) and Macrolex Fluorescent Yellow 10GN (Color Index Solvent Yellow 160:1, from Bayer Corporation). Among these, Permanent Pink R is preferred.

Any type of pigment that is well known for inclusion in thermoplastic materials can also be added to the thermoplastic matrix. Preferred pigments

include titanium dioxide, zinc sulfide, carbon black, cobalt chromate, cobalt titanate, cadmium sulfides, iron oxide, sodium aluminum sulfosilicate, sodium sulfosilicate, chrome antimony titanium rutile, nickel antimony titanium rutile, zinc oxide, and polytetrafluoroethylene.

5           It may also be advantageous to include various chemicals to prevent degradation of the thermoplastic matrix due to exposure to UV light (hereinafter "UV stabilizers"). Suitable UV stabilizers include substituted benzotriazoles, or triazines, or tetraalkylpiperidines. The UV stabilizers may be mixed into the thermoplastic matrix, or they can be included only in a  
10   "hardcoat" transparent protective layer which is applied over the viewing surface.

          The resin composition according to the invention may further contain other resins and additives such as reinforcing agents, fillers, impact modifiers, heat resisting agents, antioxidants, anti-weathering agents, stabilizers, mold  
15   release agents, lubricants, nucleating agents, plasticizers, flame retardants, flow-improving agents and anti-statics. These additives may be introduced in a mixing or molding process, provided the properties of the composition are not damaged.

          The reinforcing fillers may be metallic fillers such as fine powder  
20   aluminum, iron, nickel, or metal oxides. Non-metallic fillers include carbon filaments, silicates such as mica, aluminum silicate or clay, talc and asbestos, titanium oxide, wollastonite, novaculite, potassium titanate, titanate whiskers, glass fillers and polymer fibers or combinations thereof. Glass fillers useful for reinforcement are not particularly limited in their types or shapes and  
25   may be, for instance, glass fibers, milled glass, glass flakes and hollow or solid glass beads. Glass fillers may be subjected to surface treatment with coupling agents such as silane or titanate-type agents to enhance their adhesion with

resin, or coated with inorganic oxides to provide some surface color to the filler.

Reinforcing fillers are preferably used in an amount sufficient to yield the reinforcing effect, usually 1 to 60% by weight, preferably less than 10% by weight, based on the total weight of the composition. Glass fibers, or a combination of glass fibers with talc, mica or aluminum silicate are preferred reinforcing agents. These fibers are preferably about 0.00012 to 0.00075 inches long. The amount of filler must be less than that which would make the material opaque.

In an exemplary embodiment of the invention, a polycarbonate derived from brominated bisphenol is added as a flame retardant. When such brominated polymers are added, inorganic or organic antimony compounds may further be blended in the composition to synergistically enhance flame retardance introduced by such polycarbonate. Suitable inorganic antimony compounds are antimony oxide, antimony phosphate,  $\text{KSb(OH)}_6$ ,  $\text{NH}_4\text{SbF}_6$  and  $\text{Sb}_2\text{S}_3$ . A wide variety of organic antimony compounds may also be used, such as antimonate esters of organic acids, cyclic alkyl antimonate esters and aryl antimonate acid compounds. Examples of typical organic antimony compounds are potassium antimony tartrate, antimony salt of caproic acid,  $\text{Sb(OCH}_2\text{CH}_3)_3$ ,  $\text{Sb[OCH(CH}_3\text{)CH}_2\text{CH}_3]_3$ , antimony polymethylene glycolate and triphenyl antimony. A preferred antimony compound is antimony oxide.

Phosphites (e.g., aromatic phosphite thermal stabilizers), metal salts of phosphoric and phosphorous acid, hindered phenol antioxidants, and aromatic lactone radical scavengers may also be added as stabilizers or antioxidants.

Suitable antistatic agents include, but are not limited to, phosphonium salts, polyalkylene glycols, sulfonium salts and alkyl and aryl ammonium salts.

5        Suitable mold release agents include, but are not limited to, pentaerythritol tetracarboxylate, glycerol monocarboxylates, glycerol tricarboxylates, polyolefins, alkyl waxes and amides.

10        To prepare the resin composition of the invention, the components may be mixed by any known methods. Typically, there are two distinct mixing steps: a premixing step and a melt mixing step. In the premixing step, the dry ingredients are mixed together. This premixing step is typically performed using a tumbler mixer or a ribbon blender. However, if desired, the premix may be manufactured using a high shear mixer such as a Henschel mixer or similar high intensity device. The premixing step must be followed by a melt mixing step where the premix is melted and mixed again as a melt.

15        Alternatively, it is possible to skip the premixing step, and simply add the raw materials directly into the feed section of a melt mixing device via separate feed systems. In the melt mixing step, the ingredients are typically melt kneaded in a single screw or twin screw extruder, a Banbury mixer, a two roll mill, or similar device.

20        The composition according to present invention may then be formed into articles by any known method such as extrusion or injection molding. For example, the composition may be may be used to prepare film sheet or complex shapes via any conventional technique.

25        The thermoplastic articles according to the present invention are useful for a variety of different purposes. As some specific, non-limiting examples, they may be used for business equipment housings such as computer, monitor or printer housings, communications equipment housings such as

cellular phone enclosures, data storage device housings, appliances, or automobile parts such as instrument panel components or in a lens for a head lamp. The article can be any size or shape. Thermoplastic articles according to the invention are particularly preferred for applications where low clarity and high percent light transmission are design objectives.

The present invention is further illustrated by way of the following examples. These examples are intended to be representative of the invention and are not in any way intended to limit its scope.

#### Example 1

Six different samples were prepared to show the effect of spherical partial size versus percent light transmission, haze and clarity. Specifically, formulations were prepared as described below in Table I. All of the spherical particles were PMMA and the matrix was LEXAN® polycarbonate resin (a homopolymer based on bisphenol-A).

TABLE I

Sample #	Parts Spherical Particles*	Average Diameter Particles (microns)
1	0.53	5
2	0.53	15
3	0.53	30
4	0.53	50
5	0.53	5 <sup>+</sup>
6	0.265	5
	0.265	50

\* Based on parts per 100 part matrix resin.

\* This sample has 4x as much crosslinking agent (ethylene glycol dimethacrylate) in the PMMA as sample 1.

Each of the above six samples further comprises 0.06 parts per hundred (pph) of 2,4-di-tert-butylphenylphosphite (3:1); 0.0036 pph sodium aluminum sulfosilicate (ultramarine pigment); 0.05 pph diphenyl isodecyl phosphite (stabilizer); 0.0016 pph silicone hydride-coated TiO<sub>2</sub> (pigment); and 0.0016 pph cobalt aluminum oxide (pigment).

All of these samples were prepared by blending together the above-described ingredients and extruding them in a single or twin screw extruder with the zone temperatures set to between 480 and 550 degrees F. The pellets produced from this operation were then molded into color chips of various thickness in a Boy 15S thermoplastic molding machine set to 580 degrees F in the barrel zone, and 180 degrees F in the mold. The screw in the molding machine was operated at 200 rpm. Two sets of sample chips were molded: a first set having a uniform thickness of 0.100 inches, and a second set of two-step chips having step thicknesses of 0.062, and 0.125 inches, respectively.

Figures 1 - 3 show the results obtained upon measuring percent light transmission, haze and clarity, respectively, for these six samples. Figure 1 seems to show that transmission falls off slightly with increasing PMMA spherical particle size, but the effect is very small and could be within experimental error. In any case, the percent light transmission for these samples was relatively high (about 86 - 87%).

Figure 2 shows that haze decreases with increasing particle size to a minimum value at about 30 microns (sample #3). There is no significant further decrease from 30 microns to 50 micron (sample #4). The bimodal particle size distribution of sample 6 gives an additive effect.

Table 3 shows that the clarity increases with increasing particle size up to a maximum at a particle size of about 30 microns. There is no further increase in clarity when progressing from 30 microns to 50 micron particles. Again, sample 6 shows an additive effect.

- 5           Based on samples 5 and 1, there does not seem to be a strong dependence of any of the optical properties on the degree of PMMA cross-linking.

#### Example 2

- 10           As described above, a set of chips having three steps of different thicknesses was also prepared for samples 1-6. Figures 4 - 6, which represent combined measurements taken from both the first and second set of chips, confirm when the particle size drops below 30 microns, the clarity falls and the haze increases. Obviously, this "critical" size will vary with the chosen particles and matrix material. Figures 5 and 6 also confirm that this same  
15           trend is observed regardless of the thickness of the tested sample.

It should be remembered that the above examples merely show representative compositions according to the invention, and should not be used to limit the claimed invention in any way.

## WHAT IS CLAIMED IS:

1. A thermoplastic article which comprises a transparent matrix thermoplastic resin and suspended spherical transparent thermoplastic particles having a refractive index different from that of the matrix thermoplastic resin, wherein the article has a percent light transmission above  
5 60%, a haze of less than 95% and a clarity of greater than 70% and less than 97%, wherein the article is substantially free of any inorganic light diffusing agent.

2. The thermoplastic article according to claim 1, wherein the transparent matrix thermoplastic resin is selected from the group consisting of polycarbonates, polyetherimides, polyimides, polyamides, polyesters,  
10 polycarbonate-polyester blends, polysulfones, polyether and polyphenyl sulfones, styrene acrylonitrile, polystyrene, miscible polystyrene-polyphenylene oxide blends, acrylics, polycarbonate-polysiloxanes, polyetherimide-polysiloxanes, polyarylates, and blends and copolymers of all  
15 of the above.

3. The thermoplastic article according to claim 2, wherein the transparent matrix is selected from the group consisting of polycarbonate homopolymer or copolymers, polyester carbonates, and polyethylene terephthalate.

20 4. The thermoplastic article according to claim 2, wherein the transparent matrix thermoplastic resin is polycarbonate.

5. The thermoplastic article according to claim 3, wherein the suspended transparent thermoplastic particles are PMMA spherical particles.

6. The thermoplastic article according to claim 5, wherein the percent  
25 light transmission is above 85%.



7. The thermoplastic article according to claim 6, wherein the clarity is less than 85%.

8. The thermoplastic article according to claim 5, which comprises from 0.01 to 0.50 parts of the PMMA spherical particles per 100 parts of the transparent matrix thermoplastic resin.

9. The thermoplastic article according to claim 8, which comprises from 0.05 to 0.50 weight parts of the particles per 100 parts of the transparent matrix thermoplastic resin.

10. The thermoplastic article according to claim 5, wherein the PMMA spherical particles have an average diameter of less than 30 microns.

11. The thermoplastic article according to claim 10, wherein the PMMA spherical particles have an average diameter of less than 15 microns.

12. The thermoplastic article according to claim 11, wherein the transparent thermoplastic particles comprise a plurality of sets of PMMA spherical particles, wherein each set has a different average diameter and at least one set has an average diameter of less than 30 microns.

13. The thermoplastic article according to claim 11, wherein the PMMA spherical particles have an average diameter of more than 3 and less than 10 microns.

14. The thermoplastic article according to claim 6, which further comprises a pigment.

15. The thermoplastic article according to claim 14, wherein the pigment is selected from the group consisting of titanium dioxide, zinc sulfide, carbon black, cobalt chromate, cobalt titanate, cadmium sulfides, iron oxide, sodium aluminum sulfosilicate, sodium sulfosilicate, chrome antimony

titanium rutile, nickel antimony titanium rutile, zinc oxide, and polytetrafluoroethylene.

16. The thermoplastic article according to claim 15, wherein the pigment surface is passivated.

5        17. The thermoplastic article according to claim 16, wherein the pigment is passivated with a methyl-hydrogen silicone fluid.

18. The thermoplastic article according to claim 17, wherein the pigment is TiO<sub>2</sub>.

10       19. The thermoplastic article according to claim 6, wherein the refractive index of the polycarbonate is from 1.56 to 1.62.

20. The thermoplastic article according to claim 19, wherein the refractive index of the PMMA is from 1.46 to 1.53.

21. The thermoplastic article according to claim 20, wherein the PMMA particles have a specific gravity of about 1.10 to 1.30.

15       22. The thermoplastic article according to claim 21, wherein the PMMA is essentially 100% crosslinked.

23. The thermoplastic article according to claim 6, which further comprises 0.05 to 0.5 parts mold release agent per 100 parts of the transparent matrix thermoplastic resin.

20       24. The thermoplastic article according to claim 23, wherein the mold release agent is selected from the group consisting of pentaerythritol tetracarboxylates, glycerol monocarboxylates, glycerol tricarboxylates, polyolefins, alkyl waxes and amides.

25. The thermoplastic article according to claim 6, which further comprises 0.001 to 0.05 parts of an optical brightening agent per 100 parts of the transparent matrix thermoplastic resin.

5 26. The thermoplastic article according to claim 25, wherein the optical brightener is selected from the group consisting of aromatic stilbene derivatives, aromatic benzoxazole derivatives or aromatic stilbene benzoxazole derivatives.

27. The thermoplastic article according to claim 1, wherein the article is a business equipment housing.

10 28. A thermoplastic article which comprises a transparent matrix thermoplastic resin and spherical transparent thermoplastic particles having a refractive index different from that of the matrix thermoplastic resin, wherein the average diameter of the spherical thermoplastic resin is sufficiently small such that the clarity is below its maximum value versus average diameter of  
15 the spheres, wherein the article is substantially free of any inorganic light diffusing agent.

29. The thermoplastic article according to claim 28, wherein the haze is above its minimum value versus diameter of the spheres.

20 30. The thermoplastic article according to claim 28, wherein the transparent matrix thermoplastic resin is polycarbonate and the spherical transparent thermoplastic particles are PMMA.

25 31. The thermoplastic article according to claim 30, wherein the average diameter of the spherical particles is sufficiently small such that the clarity is less than 0.95 of its maximum value versus the diameter of the spheres.

32. The thermoplastic article according to claim 31, wherein the average diameter of the spherical particles is sufficiently small such that the clarity is less than 0.93 of its maximum value versus the diameter of the spheres.

5           33. The thermoplastic article according to claim 28, wherein the article is a business equipment housing.

10           34. A thermoplastic article which comprises a transparent matrix thermoplastic resin and spherical transparent thermoplastic particles having a refractive index different from that of the matrix thermoplastic resin, wherein the size, type and loading of the spherical transparent thermoplastic particles is selected such that the haze is below 95% and the clarity is less than 85%.

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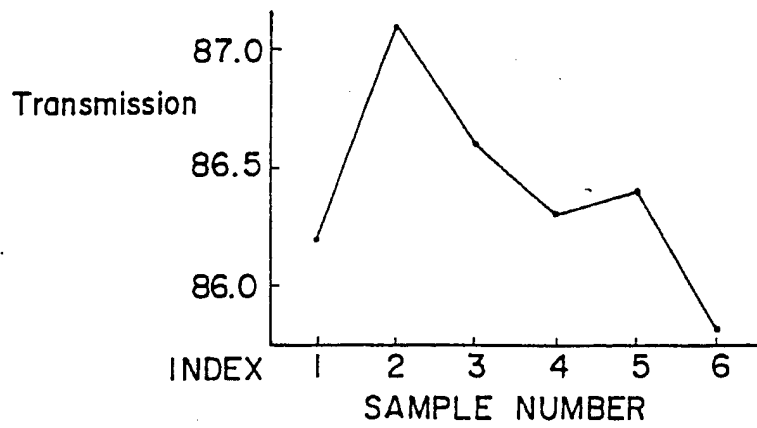


Fig. 1

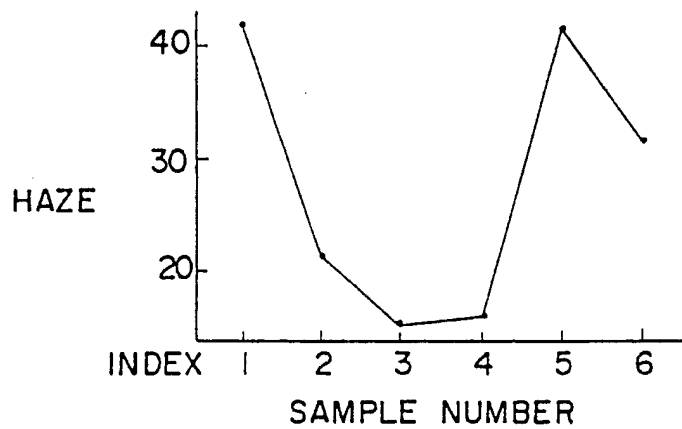


Fig. 2

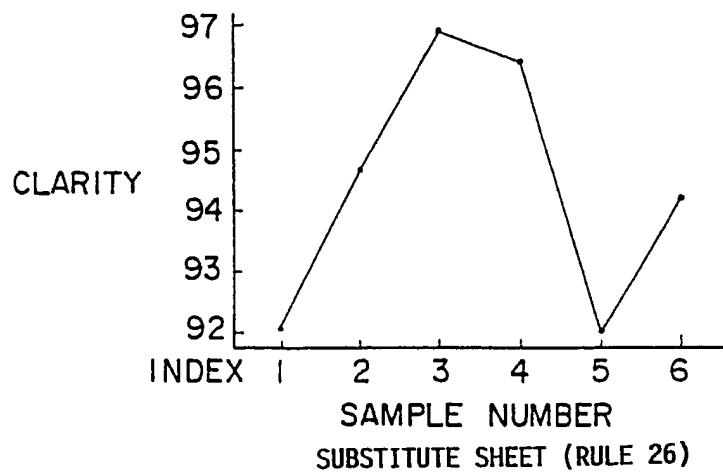


Fig. 3

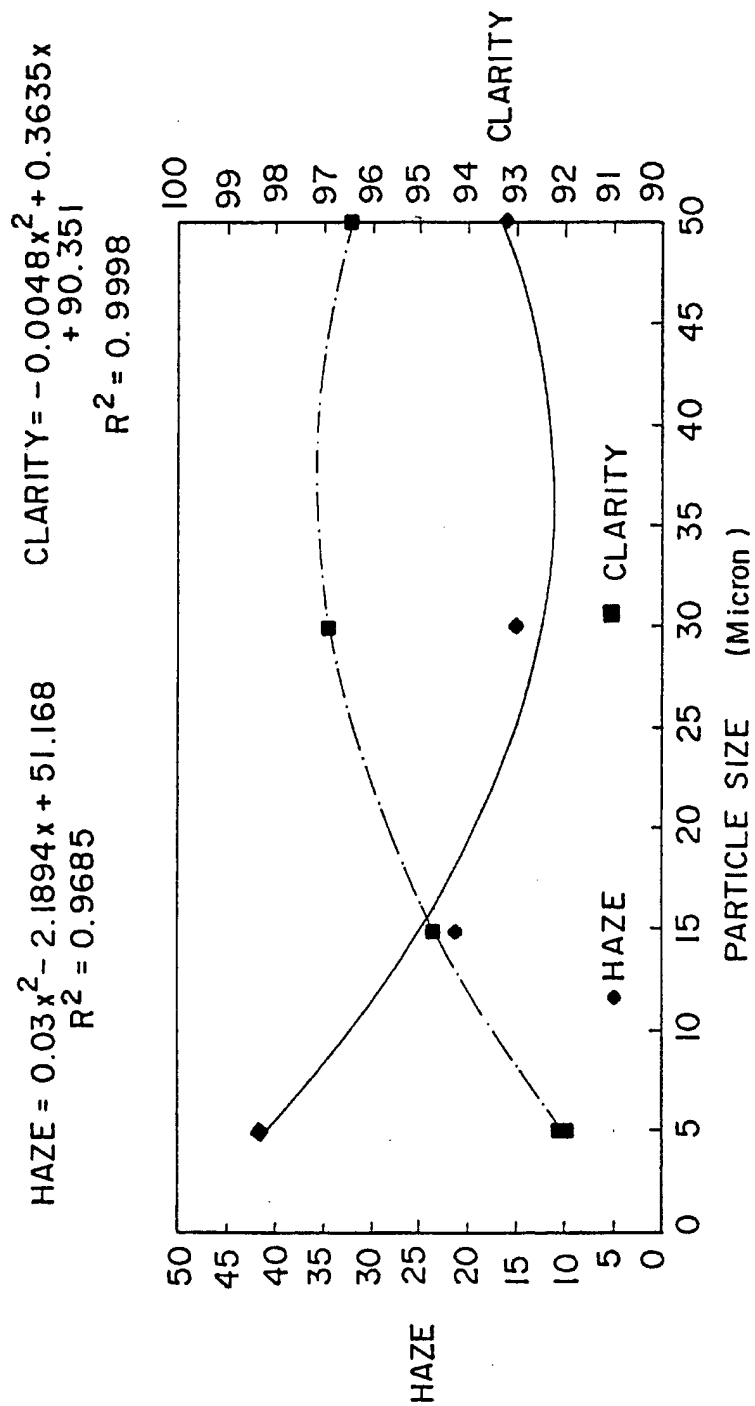


Fig. 4

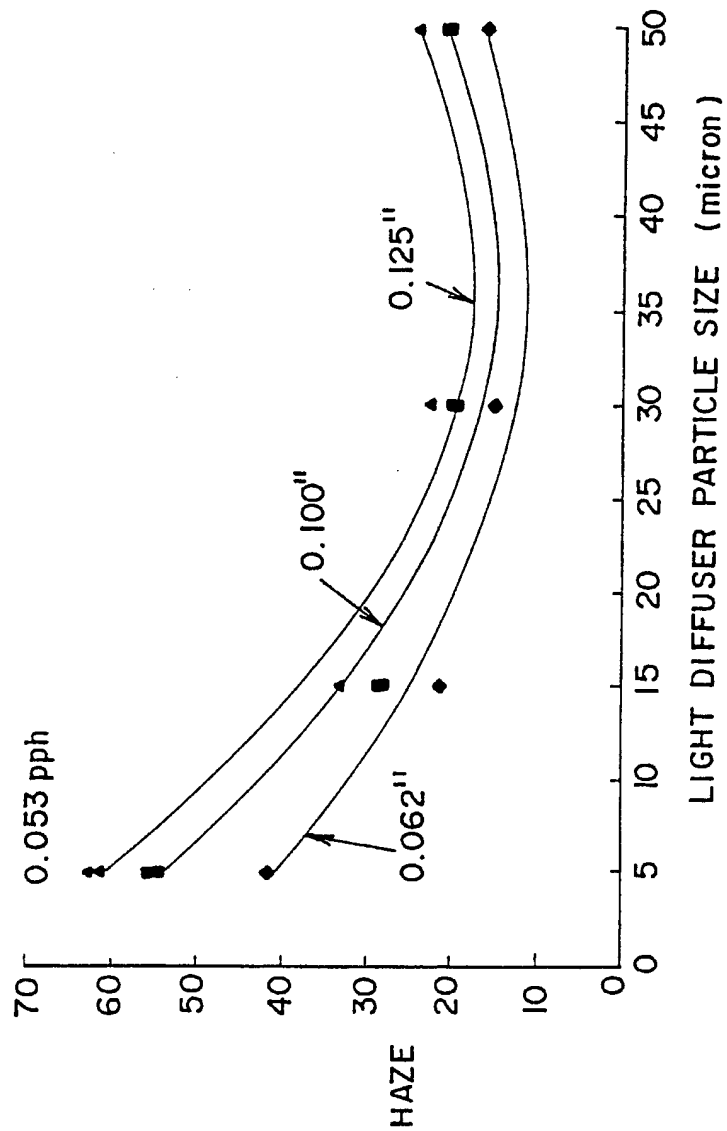


Fig. 5

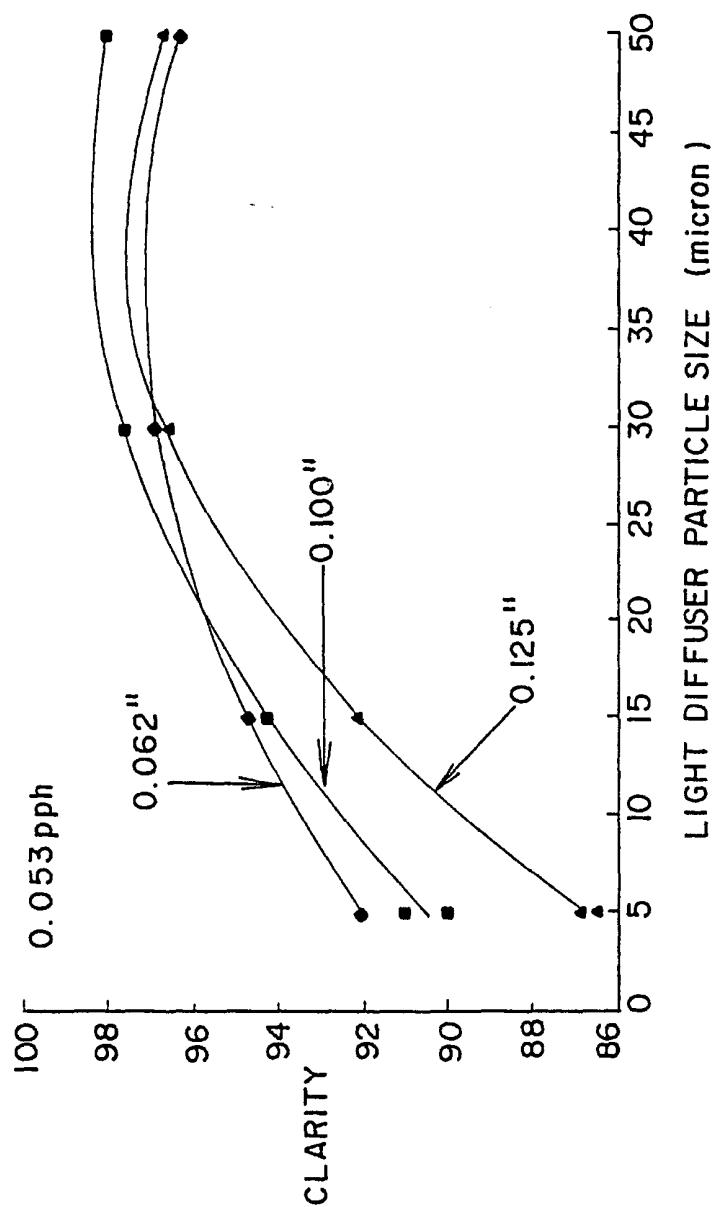


Fig. 6



# INTERNATIONAL SEARCH REPORT

International Application No

PCT/US 99/20424

**A. CLASSIFICATION OF SUBJECT MATTER**  
IPC 7 C08L101/00

According to International Patent Classification (IPC) or to both national classification and IPC

**B. FIELDS SEARCHED**

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 C08L

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the International search (name of data base and, where practical, search terms used)

**C. DOCUMENTS CONSIDERED TO BE RELEVANT**

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	CHEMICAL ABSTRACTS, vol. 116, no. 4, 27 January 1992 (1992-01-27) Columbus, Ohio, US; abstract no. 22152, XP002126370	1-13, 19-22, 27-34
Y	abstract  & JP 03 143950 A (NIPPON GE PLASTICS, LTD.) cited in the application --- -/--	1-15, 17, 20-26, 28-34

☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

\* Special categories of cited documents:

- "A" document defining the general state of the art which is not considered to be of particular relevance
- "E" earlier document but published on or after the international filing date
- "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
- "O" document referring to an oral disclosure, use, exhibition or other means
- "P" document published prior to the international filing date but later than the priority date claimed

- "T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
- "X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
- "Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.
- "&" document member of the same patent family

Date of the actual completion of the international search

21 December 1999

Date of mailing of the international search report

13/01/2000

Name and mailing address of the ISA  
European Patent Office, P.B. 5818 Patentlaan 2  
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Fax: (+31-70) 340-3016

Authorized officer

Hoepfner, W

## INTERNATIONAL SEARCH REPORT

International Application No

PCT/US 99/20424

## C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	CHEMICAL ABSTRACTS, vol. 122, no. 12, 20 March 1995 (1995-03-20) Columbus, Ohio, US; abstract no. 134965, XP002126371	1,2,28, 29,33,34
Y	abstract  & JP 06 220290 A (SEKISUI PLASTICS) cited in the application ----	1-15,17, 20-26, 28-34
Y	US 5 364 926 A (T. SAKASHITA ET AL.) 15 November 1994 (1994-11-15) cited in the application page 1, line 7 - line 17 examples claims ----	1-4, 28-34
X	CHEMICAL ABSTRACTS, vol. 117, no. 2, 13 July 1992 (1992-07-13) Columbus, Ohio, US; abstract no. 9164, XP002126380 abstract ----	1-4,28, 29,34
X	-& PATENT ABSTRACTS OF JAPAN vol. 16, no. 114 (C-0921), 23 March 1992 (1992-03-23) & JP 03 285958 A (DAINICHISEIKA COLOR AND CHEMICALS MFG. CO.) abstract ----	1-4,28, 29,34
X	EP 0 491 266 A (RÖHM GMBH) 24 June 1992 (1992-06-24) column 1, line 3 - line 5 column 6, line 14 - line 16 column 6, line 51 - line 53 ----	1-5,28, 34
Y	DATABASE WPI Week 198819 Derwent Publications Ltd., London, GB; AN 1988-128421 XP002126429 & JP 63 022816 A (ASAHI OLIN KK) abstract ----	1,14,15, 17,23
Y	EP 0 582 383 A (ROHM & HAAS) 9 February 1994 (1994-02-09) claims 1,2 ----	1,23,24
Y	EP 0 680 996 A (GENERAL ELECTRIC) 8 November 1995 (1995-11-08) page 2, line 44 - line 59 page 4, formulae I, II, III page 5, line 41 -----	1,14,25, 26

# INTERNATIONAL SEARCH REPORT

International application No.

PCT/US 99/20424

## Box I Observations where certain claims were found unsearchable (Continuation of Item 1 of first sheet)

This International Search Report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☐ Claims Nos.:  
because they relate to subject matter not required to be searched by this Authority, namely:
  
2. ☒ Claims Nos.: 1, 28, 29, 34  
because they relate to parts of the International Application that do not comply with the prescribed requirements to such an extent that no meaningful International Search can be carried out, specifically:  
see FURTHER INFORMATION sheet PCT/ISA/210
  
3. ☐ Claims Nos.:  
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

## Box II Observations where unity of invention is lacking (Continuation of Item 2 of first sheet)

This International Searching Authority found multiple inventions in this International application, as follows:

1. ☐ As all required additional search fees were timely paid by the applicant, this International Search Report covers all searchable claims.
  
2. ☐ As all searchable claims could be searched without effort justifying an additional fee, this Authority did not invite payment of any additional fee.
  
3. ☐ As only some of the required additional search fees were timely paid by the applicant, this International Search Report covers only those claims for which fees were paid, specifically claims Nos.:
  
4. ☐ No required additional search fees were timely paid by the applicant. Consequently, this International Search Report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

### Remark on Protest

- ☐ The additional search fees were accompanied by the applicant's protest.
- ☐ No protest accompanied the payment of additional search fees.

## FURTHER INFORMATION CONTINUED FROM PCT/ISA/ 210

Continuation of Box I.2

Claims Nos.: 1, 28, 29, 34

Present claims 1, 28, 29, 34 relate to a composition by reference to the following parameters: refractive index, light transmission, haze, clarity and particle size.

The use of these parameters in the present context is considered to lead to a lack of clarity within the meaning of Article 6 PCT. It is impossible to compare the parameters the Applicant has chosen to employ with what is set out in the prior art. The lack of clarity is such as to render a meaningful complete search impossible. Consequently, the search has been restricted to the subject-matter of the claims 2-27 and 30-33.

The applicant's attention is drawn to the fact that claims, or parts of claims, relating to inventions in respect of which no international search report has been established need not be the subject of an international preliminary examination (Rule 66.1(e) PCT). The applicant is advised that the EPO policy when acting as an International Preliminary Examining Authority is normally not to carry out a preliminary examination on matter which has not been searched. This is the case irrespective of whether or not the claims are amended following receipt of the search report or during any Chapter II procedure.

# INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/US 99/20424

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
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JP 6220290 A	09-08-1994	NONE	
US 5364926 A	15-11-1994	EP 0615996 A	21-09-1994
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PCT

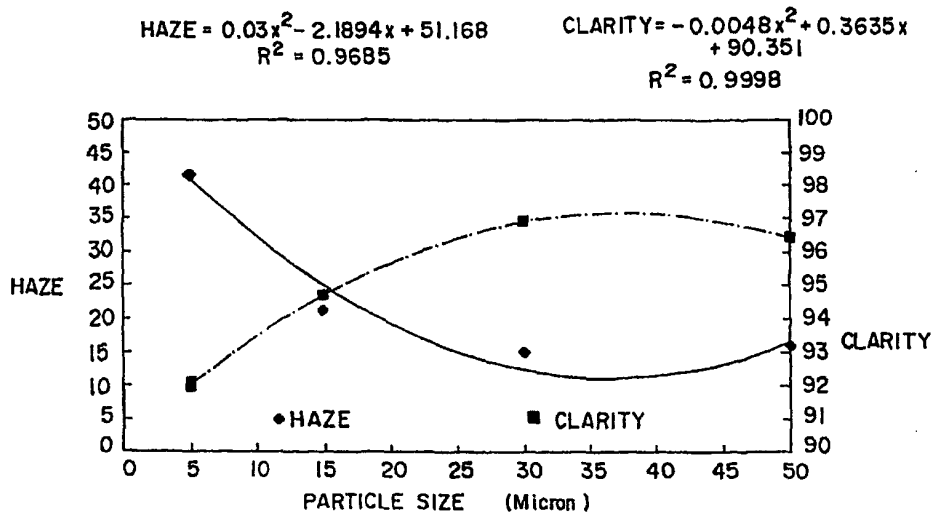
WORLD INTELLECTUAL PROPERTY ORGANIZATION  
International Bureau



INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification <sup>7</sup> : C08L 101/00		A1	(11) International Publication Number: WO 00/27927
			(43) International Publication Date: 18 May 2000 (18.05.00)
(21) International Application Number: PCT/US99/20424		(81) Designated States: CN, JP, SG, European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE).	
(22) International Filing Date: 7 September 1999 (07.09.99)			
(30) Priority Data: 09/187,637 6 November 1998 (06.11.98) US		Published With international search report.	
(71) Applicant: GENERAL ELECTRIC COMPANY [US/US]; 1 River Road, Schenectady, NY 12345 (US).			
(72) Inventors: SKARBARDONIS, John, G.; P.O. Box 228088, Charleston, SC 29423-8088 (US). SEARCY, Everett, R.; 5024 Middle Mt., Vernon Road, Evansville, IN 47712 (US). MARTIN, Stanley, Vernon; RR#1, Box 91, Cynthiana, IN 47612 (US). LOEHR, Todd, M.; 209 Barbara Drive, Evansville, IN 47712 (US).			
(74) Agents: SNYDER, Bernard et al.; General Electric Company, 3135 Easton Turnpike W3C, Fairfield, CT 06431 (US).			

(54) Title: THERMOPLASTIC ARTICLE HAVING LOW CLARITY AND LOW HAZE



(57) Abstract

There is described herein a thermoplastic article which has a high transmittance, less than about 95% haze and low clarity. If this article is placed in front of an object, the observer on the far side of the article will see the object, but not clearly.



# Chemical Abstracts Record for JP 11076934

L1 ANSWER 1 OF 1 CAPLUS COPYRIGHT 2002 ACS  
ACCESSION NUMBER: 1999:199618 CAPLUS  
DOCUMENT NUMBER: 130:253759  
TITLE: Heat-resistant nonstick decorative steel sheets and  
coatings for their manufacture  
INVENTOR(S): Mori, Koji; Hara, Taketo; Okubo, Kenichi; Koshiishi,  
Kenji  
PATENT ASSIGNEE(S): Nisshin Steel Co., Ltd., Japan  
SOURCE: Jpn. Kokai Tokkyo Koho, 6 pp.  
CODEN: JKXXAF  
DOCUMENT TYPE: Patent  
LANGUAGE: Japanese  
FAMILY ACC. NUM. COUNT: 1  
PATENT INFORMATION:

PATENT NO.	KIND	DATE	APPLICATION NO.	DATE
JP 11076934	A2	19990323	JP 1997-262956	19970911 <--

AB The steel sheets useful for processing into cooking utensils, bear an undercoat layer obtained from a mixt. of heat-resistant polymers and PTFE or/and perfluoroalkyl vinyl ether-tetrafluoroethylene copolymer (I), and a decorative topcoat layer obtained from a colored resin similar to the above layer or from PTFE or/and I. Thus, roll coating a PTFE-polyether-polysulfone mixt. contg. TiO<sub>2</sub> on a chromated steel sheet (SUS 430) to dry thickness 12 .mu.m, baking at 400.degree. for 40 s, spray coating a PTFE-polyether-polysulfone mixt. contg. carbon black on top in water-drop pattern, and baking similarly gave a decorative product with good heat resistance and interlayer adhesion.



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Dossier: 10064023

Legal Date: 07-01-2002

No.	Doccode	Number of pages
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2	LET.	2

Total number of pages: 3

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